MECHANICAL AND THERMAL PROPERTIES OF POLYPROPYLENE/ CLAY ROOF TILES WASTE (PP/CRTW) COMPOSITES

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ABSTRACT

The usage of clay roof tiles waste as a filler in composite was studied. Polypropylene (PP) composites of clay roof tiles wastes (CRTW) for low strength applications were prepared at filler contents 5%, 10%, 15%, 20%, 25% and 30%. Unfilled PP used as reference sample. The clay roof tiles wastes were prepared to achieve 45 micron in size to mix with PP. The composites were mixed using double hot roll mill mixer at temperature 190°C. The final products of the composites prepared were in a granul form which undergone granulation process after mixing. Injection moulding process also conducted to get the composites in dumbbell shape. The mechanical and thermal properties of the composites prepared were determined. Results showed that the CRTW improved the hardness properties and melt density with the increases in the filler content. On the other hand melt flow index was however observed to decrease with increases in the filler content. For the thermal properties at maximum loading of filler content it shows the maximum temperature of weight loss rate, T_{peak} and the onset decomposition temperature, T_{onset} higher compared to unfilled PP. The tensile properties for ultimate tensile strength, elongation at break, break strain percentage, maximum load and toughness were however also observed to decrease with increases in the filler content. FTIR results showed the main functional group of PP as a dominant constituent in PP/CRTW composites still remained at the wave number around 2917~2950 cm^{-1} at all filler content.

ABSTRAK

Kegunaan bahan buangan atap genting sebagai pengisi pada komposit dikaji. Komposit Polypropylene (PP) bahan buangan atap genting tanah liat disediakan pada kandungan pengisi sebanyak 5%, 10%, 15%, 20%, 25% dan 30% untuk applikasi penggunaan kekuatan rendah disediakan. PP tanpa pengisi digunakan sebagai sampel rujukan.Bahan buangan atap genting tanahliat disediakan bagi mendapatkan saiz 45 micron untuk percampuran dengan PP. Komposit dicampur menggunakan mesin pencampur panas dua pemusing pada suhu 190°C. Produk akhir bagi komposit yang disediakan berbentukgranul yang melalui proses penggranulan selepas proses pencampuran. Proses cetakan juga dilakukan bagi mendapatkan sampel komposit yang berbentuk dumbbell. Sifat mekanikal dan terma komposit yang dihasilkan ditentukan. Keputusan menunjukkan bahan buangan atap genting tanah liat menambahbaik sifat kekerasan dan ketumpatan leburan dengan pertambahan kandungan pengisi. Sebaliknya indek aliran leburan dilihat menurun dengan pertambahan kandungan pengisi.Untuk sifat terma, kandungan maksimum pengisi menunjukkan suhu maksimum kadar kehilangan berat, T_{peak} dan suhupenguraian onset, T_{onset} lebih tinggi berbanding PP tanpa pengisi. Sifat ujian tegangan untuk kekuatan tegangan puncak, pemanjangan semasa putus, peratusan terikan semasa putus, beban maksimum dan keliatan bagaimanapun dilihat menurun dengan pertambahan kandungan pengisi. Keputusan FTIR menunjukkan kumpulan fungsi utama PP sebagai juzuk dominan komposit PP/CRTW masih dikekalkan pada gelombang sekitar 2917~2950 cm^{-1} untuk semua kandungan pengisi.

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of polypropylene nano-composites

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LIST OF SYMBOLS AND ABBREVIATIONS

ATR	-	Attenuated Total Reflectance
$CaCO_3$	-	Calcium carbonate
CRTW	-	Clay roof tiles waste
Fe_2O_3	-	Iron Oxide
FTIR	-	Fourier transform infrared spectroscopy
H_s	-	Shore hardness values
MFI	-	Melt Flow Index
MMT	-	Montmorillonite
MnO_2	-	Mangan Oxide
PET	-	Polyethylene
PMCs	-	Polymer matrix composites
PP	-	Polypropylene
PP/ES	-	Polypropylene/Eggshell
PS	-	Polystyrene
PVC	-	Poly Vinyl Cloride
SSSP	-	Solid state shear pulverization
XRF	-	X-Ray Fluorescence
ZrO_2	-	Zirconium Oxide

CHAPTER 1

INTRODUCTION

1.1 Research Background

Polymer composites have been subjected to increasing interest, study and utilization for some decades. There are a lot of studies conducted in this field focusing on properties improvement of the final product. In fact with the advanced and improved technology some researchers start to develop advanced materials in nano scale known as nano composites. Polypropylene (PP)is one of most common polymer where its blends and composites has wide applications in automotive parts, home appliances, packaging industry and extruded construction profiles (Perez et al., 2013).

To enhance the mechanical properties of the final product filler plays an important role. Fillers are class of particles that are applied to polymeric composites and other materials to improve some of their properties. Elements such oxides, carbides and composite salts can be used as dispersed phase. Investigation and manufacturing of the composites obtained from fillers and common polymers including polypropylene (PP), polyethylene (PE), polyurethanes (PU), polyamides and polystyrene which nowadays become a main focus of many laboratories in polymer science (Nowicki et al., 2007).

Polymer composites with inorganics fillers such as glass fibers, silica nanoparticles, graphene, carbon nanotubes and nanofibers have been studied extensively over the past two decades. Inorganic mineral fillers like calcium carbonate (CC), silica and talc have gained interest as low cost fillers for thermoplastics. These fillers offer significant enhancement in stiffness, crystallization rate and thermal stability. There has been growing demand for green and renewable substitutes for inorganic fillers. Such fillers offer major advantages such as low density as well as reduced cost and mechanical wear during processing. Natural fibers, rice husk and wood flour are the most widely investigated green fillers (Iyer&Torkelson, 2014).

The addition of clay mineral fillers in polymers has also become very popular research interest in the last four decades to improve the mechanical, electrical, thermal, optical and processing properties of polymer. There are many kind of clay minerals such as bentonite, kaolin, talc and mica have been used as an inorganic fillers for polymer composites mainly either for cost reduction or properties enhancement such as modulus, hardness, thermal stability, electrical insulation, thickening and opacity. For example, PP-clay composites have been prepared with PP and an unmodified sodium bentonite clay of Indian origin (Sarkar et al., 2008).

1.2 Problem Statement

Nowadays with growing awareness to minimize or reduce environmental impacts throughout the world has forced researchers to synthesize new green materials and processes that enhance the environmental quality of products. The concepts of 3R which is reduce, re-use and recycle become an important considerations in the fabrications of new products. The focus is to prevent pollution. In this perspective, biodegradability, ecofriendliness, easy availability, sustainability, light weight become the main focus in the product design in most industrial and manufacturing sector. The waste management hierarchy suggests that waste should be reduced, reused, recycled then be disposed to proper dumpsite like landfill(Nagapan et al., 2013).

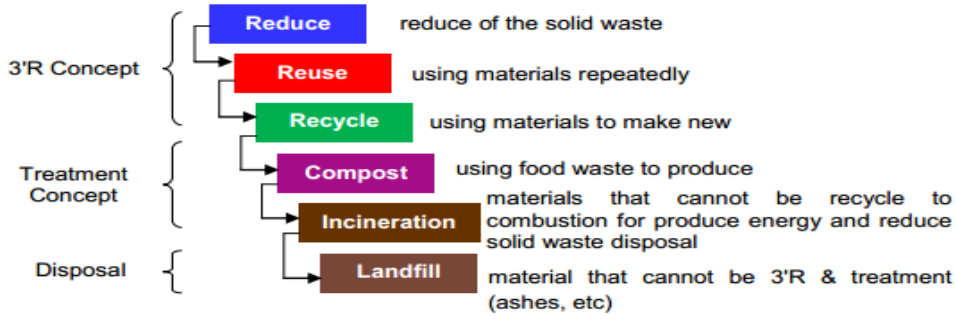

Figure 1.1: Waste management hierarchy (Nagapan et al., 2013)

Therefore to support the waste management policy a lot of studies have been conducted for recycling waste. For example a study by Iyer & Torkelson (2014) had proved that egg shells can be recycled as filler for polymer composite. Green composites of polypropylene and eggshell (PP/ES) able to produce through solid state shear pulverization (SSSP) with excellent properties achieved. A study by Halicka et al., (2012) proven that ceramic sanitary ware waste can be used as concrete aggregate with the strength higher by 2 to 8%.

Traditionally, polymeric materials have been filled with synthetic or natural inorganic compounds in order to improve their properties or to reduce cost. Polypropylene is a polymer of choice for a large number of applications. It has a low density of 0.9 g/cm^3, high crystallinity as well as low cost. The products made out of polypropylene can withstand high melting temperature up to 160°C higher than boiling water temperature owing to its higher softening point and dimensional stability. To expands the applications of PP, composites by the incorporation of inorganics fillers such as silica particles, talc and clay have been commonly used (Mittal, 2012).

In Malaysia, Figure 1.2shows that on average the generation of waste can be breakdown into 36.73% from house hold wastes, 28.34% from industrial and construction waste while other sources (market and commercial waste, institutional waste, landscaping waste and street sweeping waste) account for the remaining 34.93% in the Central and Southern region of Malaysia. However industrial waste management constitutes one of the major problems in today society. Recycling of non-biodegredable waste is particularly difficult. Clay based material is one of the materials that has been widely used in construction materials instead of wood, sand,

concrete and other waste materials. This material is a major compound in clay brick, clay tiles and clay roofing tiles due to its wide-ranging properties, highresistance to atmospheric condition, geochemical purity, and easy access to its deposits near the earth's surface and low price(Seen, 2009).

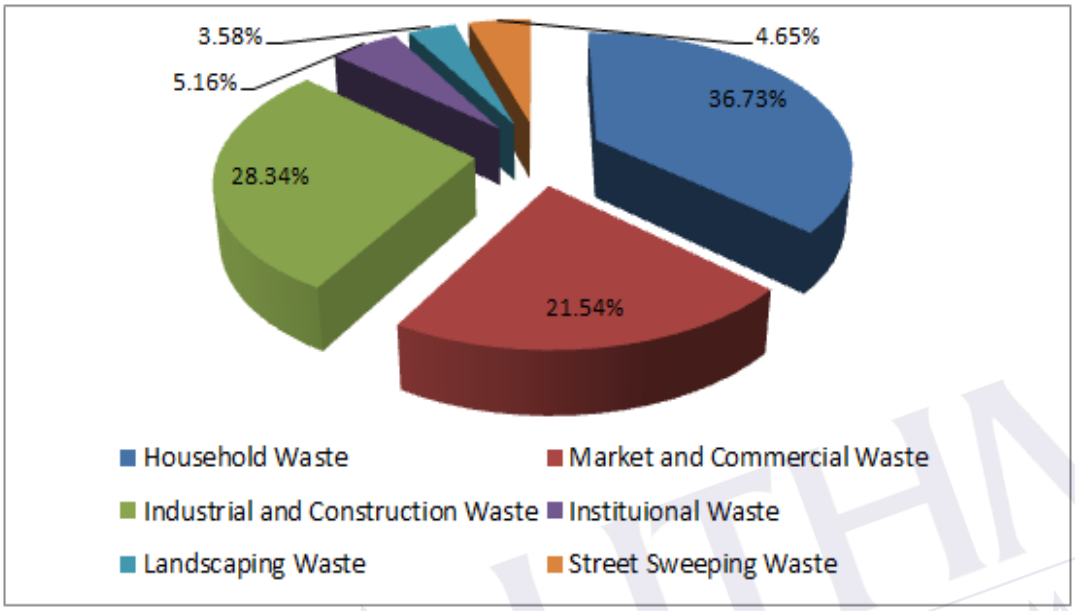

Figure 1.2: Percentages of different types of solid waste generated (tonne/day) in Malaysia (Seen,2009)

Ceramic waste either from construction or manufacturing has been classified in this category due to prolonged biodegradation period of ceramic (up to 4 thousand years). Ceramic industry waste constitutes significant share in the total production, thus recycling a ceramic waste is a big issue. For the cost saving reasons mainly on the machineries and maintainence most of the ceramic manufacturer either those making a floor tiles, sanitary ware products, or clay roof tiles preferred not to re-cycle or re-use the fired clay for their final products or re-production. Another main concern is related to their properties of the final products. Recycled materials will not give better mechanical properties if the ratios with the virgin raw materials not proper study.

Based on the materials used for its production, ceramic ware can be divided into two groups. The first group included products of burned red clay such as bricks, structural wall and floor tiles and also roof tiles after gone through high temperature of firing up to 1000°C. Another group are products made of white clay included

technical ceramics used as electrical insulators, ceramic sanitary ware (washbowls, lavatory pands, bathtubs), medical and laboratory vessels (Halicka et al., 2013). Industrial ceramic wastes including clay roof tiles wastes (CRTW) usually come from defectives products or roof tiles rejected during quality control. Main defects include cracks, glaze defects, nicks, broken and crazing.

One of the natural ways of reusing inorganic industrial wastes, is their use in the production of building materials, especially as raw materials in the concrete manufacturing. This manner of recycling has positive impact on the environment because it reduce the amount of deposited waste. Anyway, there are a number of research works on the properties of clay reinforced polymer composites. However, limited study on the properties of clay waste used as filler in polymeric materials which is in this case CRTW. In this project CRTW will be used as filler to produce PP/CRTW polymer composites. If compared to other clays such as bentonite, talc, mica and kaolin which mostly used in a lot of research in a virgin condition with enhancement in the surface modification.

Therefore due to this reasons since this study never conducted yet the important characterization of this PP/CRTW not exactly known. This research is very good to be conducted to analyze the microstructure, chemical composition, mechanical and thermal properties from composites of PP and CRTW. Figure 1.3 shows the CRTW at Terreal (M) Sdn. Bhd which rejected mainly due to process reject.

Figure 1.3: Clay roof tiles waste at Terreal (M) Sdn. Bhd image recorded on 31st Dec 2014

1.3 Research Objectives

This study embarks on the following objectives:

1) To fabricate PP composites using CRTW with below than 45 micron in particles as filler for low strength applications at different weight of CRTW content in the range of 5%, 10%, 15%, 20%, 25% and 30%.
2) To determine the mechanical and thermal properties of the fabricated PP/CRTW composites for low strength applications and compared the results with unfilled PP without any surface modification conducted.

1.4 Scope of Study

This study will cover the followings:

1) Preparation of CRTW in which the particle size below than 45 micron.

2) Preparation of PP with CRTW based on the specified weight composition in different ratios through blending process.

3) Only PP and CRTW were mixed together without any binder/compatibilizer. No surface modification conducted.

4) Test and analyse the properties of the samples including particle size, chemical and material composition, mechanical and thermal properties based on the material standard.

CHAPTER 2

LITERATURE REVIEW

2.1 Introduction

The expanding industrial activities create a continual demand for improved materials that satisfy increasingly stringent requirements, such as high strength, modulus, thermal and/or electrical conductivity, heat distortion temperature, lower thermal expansion coefficient and reduced cost. Many of modern technologies recently require materials with unusual combinations of properties that cannot be met by the conventional metal alloys, ceramics and polymeric materials. These requirements, which often involve a combination of many difficult to attain properties, may dictate the use of composite material whose constituents will act synergistically to solve the needs of the application (Callister, 2004).

Many composites used today are at the leading edge of materials technology, with performance and costs appropriate to ultrademanding applications. But heterogeneous materials combining the best aspects of dissimilar constituents have been used by nature for millions of years. There are extensively used as materials in making aircraft structures, electronic packaging to medical equipment, and space vehichle to home building (Roylance,2008).

As composites offer unusual combinations of component materials properties such as weight, strength, stiffness, permeability, electrical, biodegradability and

optical properties that is difficult to attain separately by individual component thus a composite material can be custom tailored to have specific properties that will meet special requirements. The optimum design of composite materials and manufacturing process to meet the target properties for specific engineering application is very important (Olad, 2008).

The use of composites in the aerospace industry has increased since 1970s. Traditional materials for aircraft construction include aluminum, steel and titanium. The performance advantages associated with reducing the weight of aircraft structural elements has been the major focus for military aviation composites development. The same approaches also goes in other sectors such as submarine and automotive. Materials experts from various automakers estimate that advanced composite auto body could be 50-70% lighter than a current similarly sized steel auto body as compared with a 40-55% mass reduction for an aluminum auto body and a 25-30% mass reduction for an optimized steel auto body. The diversity of properties belonging to composite materials has made this field more interesting to be explored (Koronis,2013).

2.2 Composite Materials

Composites can be defined as materials that consist of two or more chemically and physically different phases separated by a distinct interface. The different systems are combined to achieve a system with more useful structural or functional properties non attainable by any of the constituent alone. The basic difference between blends and composites is that the two main constituents in the composites remain recognizable while these may not be recognizable in blends (Jose et al.,2012).

Generally composite material consist of one or more discontinuous or dispersed phase distributed in one continous phase. In the case of several discontinuos phases of different natures the composites is called a hybrid composites. Usually the discontinuos phase is harder than the continuos phase with better mechanical properties. The continuos phase is called the matrix which usually more ductile and less hard. It holds the dispersed phase and shares a load with it. Figure 2.1 illustrate the constituents of composite consists of the discontinuos phase called

as reinforcement material or filler (Barthelot, 1999). Fillers are class of particles that are applied to composite materials to improve some of their properties. It is embedded in the matrix in a discontinuos form.

The properties of composites are a function of the properties of the constituent phases, their relative amounts, and the geometry of the dispersed phase. "Dispersed phase geometry" in this context means the shape of the particles, particle size, distribution, and orientation. Figure 2.2 and Figure 2.3 show the classification of composites material which consist of three main divisions: particle- reinforced, fiber- reinforced, and structural composites; also at least two subdivisions exist for each. The dispersed phase for particle-reinforced composites is equiaxed (i.e., particle dimensions are approximately the same in all directions); for fiber-reinforced composites, the dispersed phase has the geometry of a fiber (i.e., a large length-to-diameter ratio). Structural composites are combinations of composites and homogenous materials show the types of composites based on the form of reinforcement (Callister, 2004).

The reinforcement in the matrix can be divided into polymeric, ceramic and metallic composites. Polymers often have advantages over other materials such as metals and ceramics. There are widely used in various technical applications because of their unique advantages such as ease of production, light weight and ductile. However polymers have lower mechanical modulus and strength properties compared to metals and ceramics. The commercial importance of polymers and their increasing use, results to the continuos demand for improvement in their properties to meet the necessary conditions. By the composite technology, polymer properties are improved while maintaining their light weight and ductile nature (Olad, 2008).

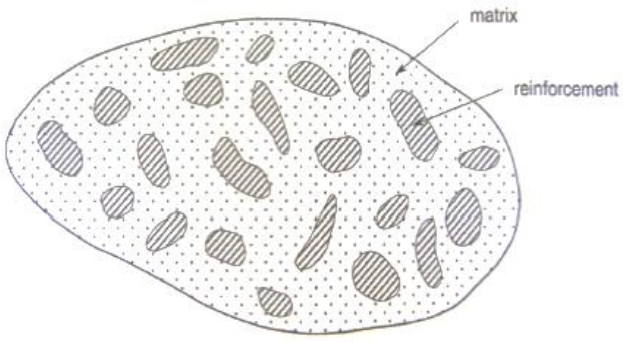

Figure 2.1: Composite materials (Barthelot,1999)

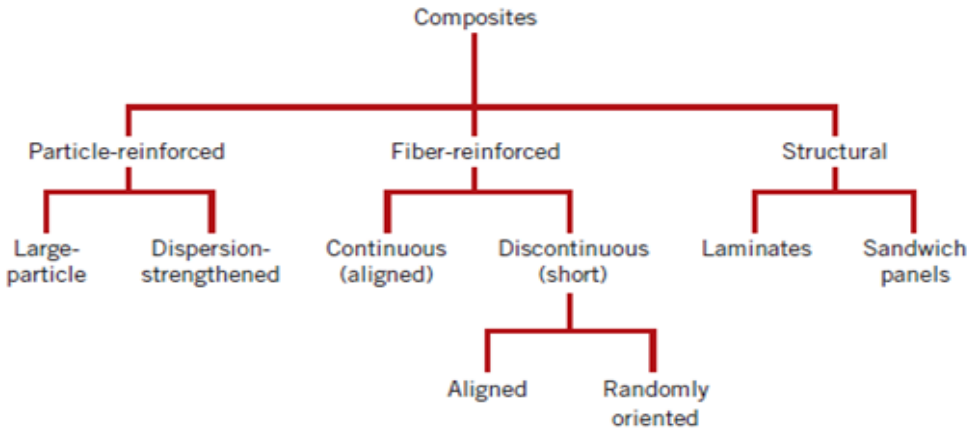

Figure 2.2: Types of reinforced composites (Callister, 2004)

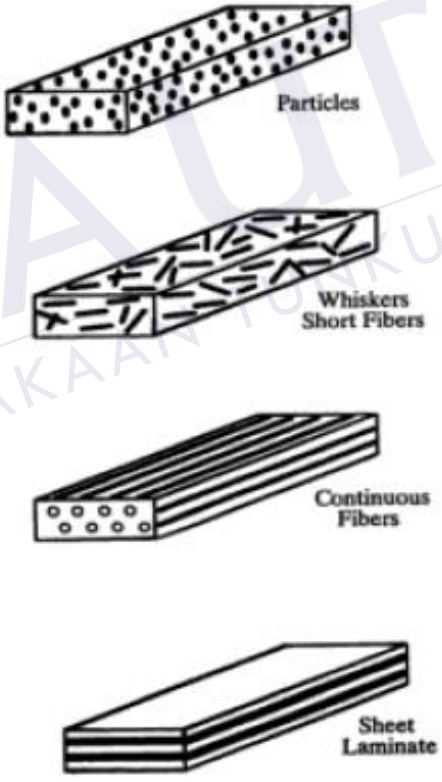

Figure 2.3: Types of composites based on the form of reinforcement (Chawla, 2013)

2.3 Classification of composite materials based on matrix

On the basis of matrix phase, composites can be classified into metal matrix composites, ceramic matrix composites and polymer matrix composites Figure 2.4 (Jose et al.,2012).

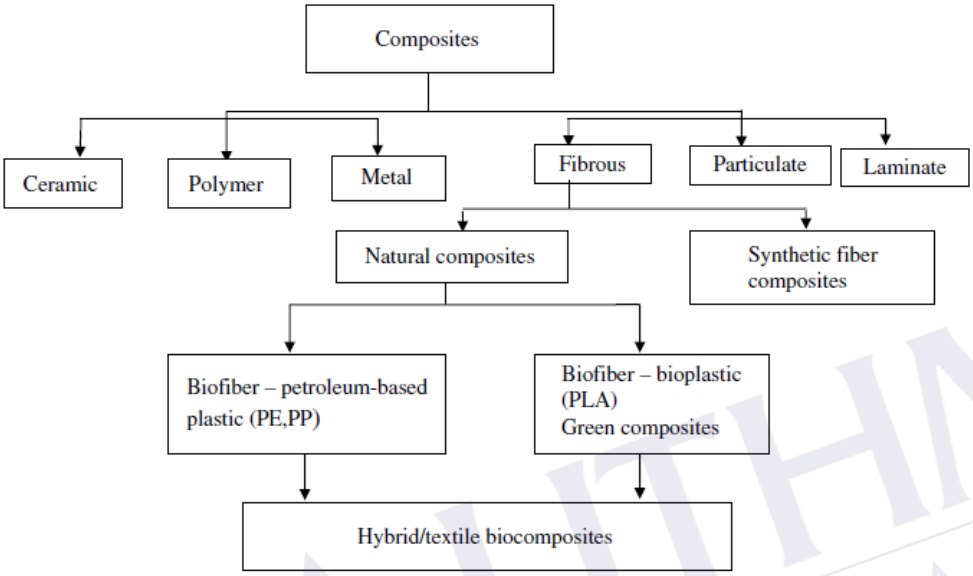

Figure 2.4: Classification of composites (Jose et al., 2012)

2.3.1 Polymer Matrix Composites (PMCs)

A polymer composite is made by the combination of a polymer and synthetic or natural inorganic filler. Fillers are employed to improve the desired properties of the polymer indirectly reduce the cost. Nowadays, the application of polymer composites as engineering has become state or the art. Polymer composites been developed with improved mechanical, thermal, barrier and fire retardancy properties are used in various applications (Olad, 2008).

Therefore the efficiency of properties improvement strongly depends on the properties of the choosen filler. The influence factors included the mechanical properties of the filler, adhesion between matrix and filler as well as the ratio of the filler. To optimize the composite properties the most essential factor that leads this is a good dispersion (intercalation and exfoliation) and also the distribution of the filler in the matrix (Olalekan et al., 2010). Important types of modified polymer systems

include polymer composites, polymer-polymer blends and polymeric foams. In general, parameters affecting the properties of polymer composites, include (Xanthos, 2010):

- The properties of the additives (inherent properties, size, shape)
- Composition
- The interaction of components at the phase boundaries, which is also associated with the existence of a thick interphase; this is often considered as a separate phase, controlling adhesion between the components
- The method of fabrication

Today polymeric composites market demand in industry had increased significantly as substitute materials for metals in applications within the aerospace, automotive, marine, sporting goods and electronic industries. Their light weight and superior mechanical properties make them especially suited for transportation applications.

2.3.2 Polymer Matrices

The matrices in composite materials have the role of transferring the mechanical loading to the fillers and to protect them from the outside environment. Therefore the matrices usually flexible and offer good compatibility with the fillers. In addition they must have a low density to keep in the composites's high specific mechanical characteristics (Berthelot, 1999). Polymers are used as the matrix phase for the composites namely polymer matrix composites. The polymer matrices are divided into two types, thermosetting resins and thermoplastic resins. These two types of matrix have the property of being able to be molded or manufactured in order to yeild either a finished product or a semi finished product whose form can be modified (Berthelot, 1999).

2.3.2.1 Thermosetting Resins

In theory, thermosetting polymers become permanently hard when heat is applied and do not soften upon subsequent heating. During the initial heat treatment, covalent crosslinks are formed between adjacent molecular chains; these bonds strengthen the chains together to resist the vibrational and rotational chain motions at high temperatures. Thermosets have a high density of cross-links and thus only heating to excessive temperatures will cause severence of these crosslinks bonds and polymer degredation. In term of mechanical properties, thermosets are harder and stronger than thermoplastics and have better dimensional stability (Callister, 2004).

Examples of thermosetting resins include polyester resins, vinyl ester resin, epoxies, phenolic and urethane. However the most common thermosetting resin used today is polyester resin, vinyl ester and epoxy. Thermosetting resins are popular because at room temperature they are in a liquid state. This allows for convenient impregnation of reinforcing fibers such as fiberglass, carbon fiber or Kevlar.

Since thermosets resins exhibits the liquid properties at room temperature it is easy to work with. Laminators can easily remove all air during manufacturing and it also allows the ability for rapid manufacturing. Beyond ease of manufacturing, thermosetting resin raw material cost also cheaper which can exhibit excellent properties. Properties of thermoset resins such as excellent resistance to solvents and corrosives, resistance to heat and high temperature, high fatigue strength, excellent adhesion and excellent finishing.

In a thermoset resin, the raw uncured resin molecules are crossed linked through a catalytic chemical reaction which is an exothermic process. The resin creates extremely strong bonds to one another and the state change from liquid to a solid. Thermosetting resin has a few disadvantages such as it cannot be reversed, reshaped, reformed or remoleded to its original condition once it catalyzed. In other words, thermoset composites are difficult to be recycled (Johnson, 2014).

2.3.2.2 Thermoplastic Resins

Almost 85% products produced worldwide are made of thermoplastic. Thermoplastic soften when heated (and eventually liquefy) and harden when cooled. It is relatively soft. Most commonly unreinforced which means it is formed into shape and have no reinforcement providing strength. Examples of thermoplastic used in industry today include Polyethylene (PET), Polypropylene (PP), Polystyrene (PS) and Polyvinyl Cloride (PVC). It consists of linear or branched chain molecules having strong intramolecular bonds but weak intermolecular bonds (Jose et al.,2012).

In general, thermoplastics materials areductile and tougher than thermoset materials. It has been used widely in various of non structural applications. Thermoplastics can be remelted by heating and solidified by cooling. In other words thermoplastics can be recycled, reshaped and reformed. Most linear polymers and those having some branched structures with flexible chains are thermoplastics (Callister, 2004).

The main reason that make thermoplastic important in our daily life it is easy to be processed. Thermoplastic composites in room temperature are in solid state but when heated a physical change occurs without required any chemical reactions as thermoset. This ability is not possible with thermosetting resins. Besides, the properties of thermoplastics also attractive. It has high delamination resistance and damage tolerance, low moisture absorption and excellent chemical resistance. Related to environmental concerns thermoplastic has very low toxicity since it does not contain any reactive chemical.

On the other hand, because thermoplastic resin is naturally in a solid state, it is much more difficult to imprenate reinforcing fiber. The resin must be heated to the melting point, pressure is required to impregnater fibers, and the composite must then be cooled under this pressure. This is complex and far different from thermoset composite manufacturing. Special tooling, technique and equipment must be used, many of which is expensive. This is the major disadvantage of thermoplastc composites. Table 2.1 summarized the advantages and disadvantages of thermosets and thermoplastics as composite matrix resin (Johnson, 2014)

Table 2.1: Advantages and disadvantages of thermosets and thermoplastics (Johnson, 2014)

Property	Thermosets	Thermoplastics
Formulations	Complex	Simple
Melt viscosity	Very low	High
Fiber impregnation	Easy	Difficult
Prepreg tac	Good	None
Prepreg drape	Good	None to fair
Prepreg stability	Poor	Excellent
Processing cycle	Long	Short to long
Processing temperature/ pressure	Low to moderate high	High
Fabrication cost	High	Low (potentially)
Mechanical properties		
-54 to 93°C, hot/wet	Fair to good	Fair to good
Environmental durability	Good	Unknown
Solvent resistance	Excellent	Poor to good
Damage tolerance	Poor to excellent	Fair to good
Database	Very large	Small

2.4 Polypropylene

Polypropylene (PP) is a semi-crystalline engineering thermoplastics and is known for its balance of strength, modulus and chemical resistance. PP has many potential applications in automobiles, appliances and other commercial products in which creep resistance, stiffness and some toughness are demanded in addition to weight and cost savings. However pure PP resin cannot meet the industrial requirements due to its low mechanical properties especially toughness and low thermal resistance. Therefore fabrication of composites is needed with addition of reinforced material or filler to improve and enhance the mechanical properties of virgin PP. Therefore PP received attention for production of polymer composites as thermoplastic matrix(Gabr et al., 2014).

PP was first product produced by Giulio Natta, following the work of Karl Zeigler, by the polymerization of propylene monomer in 1954 (Figure 2.5). The macromolecules of PP contains 10,000 to 20,000 monomer units. The stearic arrangement of methyl groups attached to every second carbon atom in the chain may vary (Figure 2.6), if all the methyl groups are on the same side of the winding spiral chain molecule, the product is referred to as isotactic PP. A PP structure where pendant methylene group are attached to the polymer backbone chain in an alternating manner is known as syndiotactic PP. The structure where pendant groups are located in a random manner on the polymer backbone is atactic form (Billmayer, 1984).

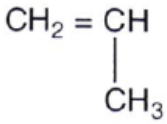

Figure 2.5:Propylene Monomer

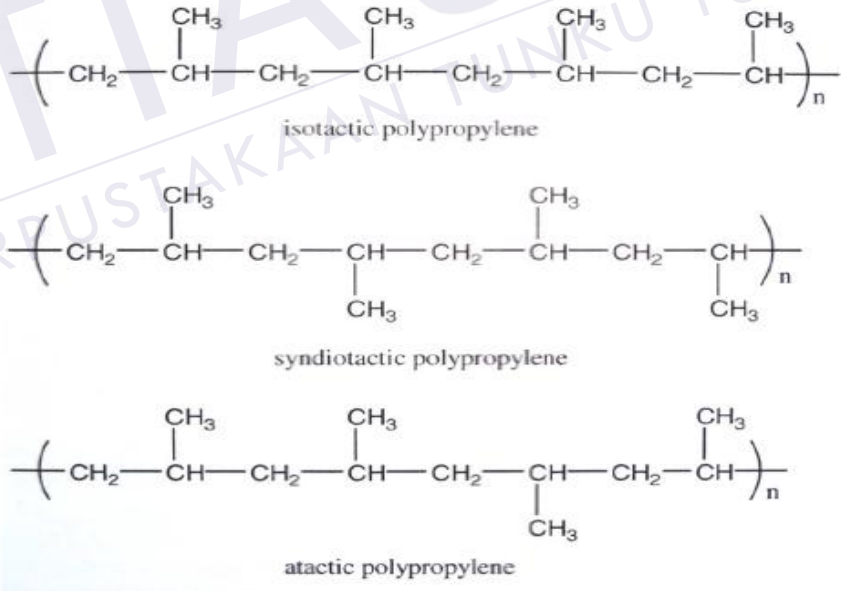

Figure 2.6: PP polymer molecules in isotactic, syndiotactic & atactic

The properties of polypropylene established by International Association of Plastic Distribution (IAPD) based on ISO and ASTM standard illustrated in the table below:

Table 2.2: Typical Properties of Polypropylene by International Association of Plastic Distribution (IAPD)

ISO or UL Test	Property	HPP*	HPP-filled	CPP**	CPP-filled
ISO1183	Specific gravity	0.90-0.91	0.97-1.27	0.89-0.91	0.98-1.24
ISO62	Water absorption (%)	0.01-0.03	0.01-0.09	0.03	0.01-0.02
ISO527	Tensile strength (MPa)	31.03-41.37	24.13-110.32	27.58-37.92	17.24-68.95
ISO527	Elongation at break (%)	100-600	1.5-80	200-500	2.2-50
ISO527	Tensile Modulus (MPa)	113.7-155.1	258.5-689.5	89.6-124.1	34.4-241.3
ISO178	Flexural Modulus (MPa)	117.2-172.3	144.8-689.5	89.6-137.9	144.8-661.9
ISO180	Notched Izod Impact Strength (J/m)	21-75	32-641	59-747	32-214
ASTM D785	Hardness, Rockwell R	80-102	75-117	65-96	81-105
ISO8302	Thermal conductivity (W/mK)	0.22	0.25-0.51	0.22	0.25-0.51
ISO11359	Coefficient of thermal expansion (10^{-4} m/m-°C)	1.4-1.8	0.27-0.90	1.08-1.80	0.36-1.08

* Homopolymer polypropylene

** Copolymer polypropylene

ASTM or UL Test	Property	HPP*	HPP-filled	CPP**	CPP-filled
D792	Specific gravity	0.90-0.91	0.97-1.27	0.89-0.91	0.98-1.24
D570	Water absorption (%)	0.01-0.03	0.01-0.09	0.03	0.01-0.02
D638	Tensile strength (psi)	4500-6000	3500-16000	4000-5500	2500-10000
D638	Elongation at break (%)	100-600	1.5-80	200-500	2.2-50
D638	Tensile Modulus (psi)	16500-22500	37500-100000	13000-18000	5000-35000
D790	Flexural Modulus (psi)	17000-25000	21000-100000	13000-20000	21000-96000
D256	Notched Izod Impact Strength (ft-ib/in of notch)	0.4-1.4	0.6-12	1.1-14	0.6-4.0
D785	Hardness, Rockwell R	80-102	75-117	65-96	81-105
C177	Thermal conductivity (10^{-4} cal-cm/sec-cm^2.°C)	2.8	2.4-9	3.5-4	3-9
D696	Coefficient of thermal expansion (10^{-5} in/in-°C)	8-10	1.5-5	6-10	2-6

* Homopolymer polypropylene

** Copolymer polypropylene

2.5 Fillers and Reinforcements

Traditionally most fillers are considered additives. An inert foreign substance added to a polymer to improve or modify its properties simplified the definition of filler (Callister, 2004). Fillers-reinforced polymer composites have gained much attention to scientific investigators in recent years. The main reason is because the incorporation of fillers alters the structural, mechanical, thermal and electrical properties of the composite materials. The degree of dispersion of filler into the polymer matrix and the interaction between filler and polymer are the main factors that affected the properties of the composite. In this context, inorganic fillers play important role in changing thermal, mechanical and electrical properties of plastics. Commonly used fillers in PP are talc, calcium carbonate and clay (Mina et al., 2009).

Other than to improve the properties of polymer, the major contribution of filler was in lowering the cost of materials by replacing the most expensive polymer, other possible economic advantages were faster moulding cycles as a result of increased thermal conductivity and fewer rejected parts due to warpage. Depending on the type of filler, other polymer properties could be affected for example melt viscosity could be significantly increased through the incorporation of fibrous materials (Xanthos, 2010).

Fillers are generally characterized based on their physical parameters like shape, size and their reinforcing effect when dispersed into continuous polymer matrix. These physical parameters could be defined in terms of aspect ratio, packing fraction and surface area. The aspect ratio is described as a ratio of themajor to the minor dimension and it defines the load transfer efficiency from the polymer matrix and the continuous phase to the filler. The packing fractions refer to the volume occupied by solid particles over the total volume. It is correlated to the aspect ratio by an inverse relationship which is leads to the concept the higher the aspect ratio, the lower the packing fraction. This implies that the fillers exhibiting high reinforcing effect must have a low packing fraction or high aspect ratio (Olalekan et al., 2010).

Fillers may be classified as inorganic or organic substance either based on the chemical family or shape depending on the size and ratio. Table 2.3 shows the fillers

classification based on chemical family and their functions summarized in Table 2.4 (Xanthos, 2010):

Table 2.3: Chemical families of fillers (Xanthos, 2010)

Chemical family	Examples
Inorganics	
Oxides	Glass (fibers, spheres, hollow spheres, and flakes), MgO, SiO_2, Sb_2O_3, Al_2O_3, and ZnO
Hydroxides	$Al(OH)_3$ and $Mg(OH)_2$
Salts	$CaCO_3$, $BaSO_4$, $CaSO_4$, phosphates, and hydrotalcite
Silicates	Talc, mica, kaolin, wollastonite, montmorillonite, feldspar, and asbestos
Metals	Boron and steel
Organics	
Carbon, graphite	Carbon fibers, graphite fibers and flakes, carbon nanotubes, and carbon black
Natural polymers	Cellulose fibers, wood flour and fibers, flax, cotton, sisal, and starch
Synthetic polymers	Polyamide, polyester, aramid, and polyvinyl alcohol fibers

Table 2.4: Fillers and their functions (Xanthos, 2010)

Primary function	Examples of fillers	Additional functions	Examples of fillers
Modification of mechanical properties	High aspect ratio: glass fibers, mica, nanoclays, carbon nanotubes, carbon/graphite fibers, and aramid/synthetic/natural fibers Low aspect ratio: talc, $CaCO_3$, kaolin, wood flour, wollastonite, and glass spheres	Control of permeability	Reduced permeability: impermeable plate-like fillers: mica, talc, nanoclays, glass flakes Enhanced permeability: stress concentrators for inducing porosity: $CaCO_3$ and dispersed polymers
Enhancement of fire retardancy	Hydrated fillers: $Al(OH)_3$ and $Mg(OH)_2$	Bioactivity	Bone regeneration: hydroxyapatite, tricalcium phosphate, and silicate glasses
Modification of electrical and magnetic properties	Conductive, nonconductive, and ferromagnetic: metals, carbon fiber, carbon black, and mica	Degradability	Organic fillers: starch and cellulosic fibers
Modification of surface properties	Antiblock, lubricating: silica, $CaCO_3$, PTFE, MoS_2, and graphite	Radiation absorption	Metal particles, lead oxide, and leaded glass
Enhancement of processability	Thixotropic, antisag, thickeners, and acid scavengers: colloidal silica, bentonite, and hydrotalcite	Improved dimensional stability	Isotropic shrinkage and reduced warpage: particulate fillers, glass beads, and mica
		Modification of optical properties	Nucleators, clarifiers, and iridescent pigments: fine particulates and mica/pigment hybrids
		Control of damping	Flake fillers, glass, and $BaSO_4$

2.6 Ceramics

Normally ceramics will mostly be reflected brittleproducts such as potteries, table wares, sanitary wares, floor tiles, roof tiles, bricks and glass. The word ceramic is originated from Greek word *keramikos* which means burnt stuff. This indicates that desirable properties of ceramics are normally achieved through a high temperature heat treatment process called firing. A ceramic is inorganic, non metallic solid comprising metal, non metal atoms which held in ionic and covalent bonds. Because ceramics are composed of at least two elements, and often more, their crystal structures are more complex than metals. The atomic bonding of ceramic itself ranges from purely ionic to totally covalent. Usually ceramic products are divided into four categories (Callister,2004):

- Structural – Mainly used for building construction such as bricks, clay pipes, floor and roof tiles.
- Refractory –Mainly used as kiln linings, ceramic fiber blanket that can withstand the temperature above 1000◦C.
- White wares – Including table wares, cook wares, sanitary wares, wall tiles and pottery products.
- Advanced ceramics – Special fine ceramics used in medical applications such as biomedical implants, coatings and electrical insulators.

2.6.1 Clay Products

One of the most widely used ceramic raw materials is clay. This inexpensive ingredient found naturally in great abundance, often is used as mined without any upgrading of quality. Another reason that makes clay becomes popular because it can be formed easily. With proper proportions during mixing, clay and water form a plastic mass that is very amenable to shaping. The formed piece is dried to remove some of the moisture, after which it is fired at an elevated temperature to improve its mechanical strength.

Most of the clay based products fall within two broad classifications which are structural clay products and the white wares. Structural clay products include building bricks, tiles, and sewer pipes. The white wares ceramics become white after

high temperature firing. Included in this group are porcelain, pottery, tableware, china wares and also sanitary wares. In addition to clay, many of these products also contain non plastic ingredients which influence the changes that take place during the drying and firing processes and the characteristics of the finished piece (Askeland, 2006).

2.6.1.1 Clay Roof Tiles

Clay roof tile as shown in Figure 2.7 is a type of the burned red clay in ceramic ware group (Halicka et al., 2013). In industry hydraulic pressing is widely adopted for the shaping of tiles. Modern hydraulic presses can provide high compaction force, high productivity, consistency and easy adjustment. Many presses are now fitted with electronic control units, which can check the height of units and automatically adjust the cycle to ensure size uniformity. For clay roof tiles it usually formed by pressing plastic clay 'bats' cut from an extruded column. The manufacturing process of clay roof tiles goes through the stages of mining or quarrying of raw materials, storage raw materials, raw materials preparation, shaping, drying, firing and subsequent treatment. Special requirements for the surface and colour of the products involve a surface treatment by glazing, engobing or profiling. The following Figure 2.8 shows a basic schematic view of clay roof tiles manufacturing process (European Commission, 2007).

Figure 2.7: Clay Roof Tiles adopted from Texas-Clay Roof

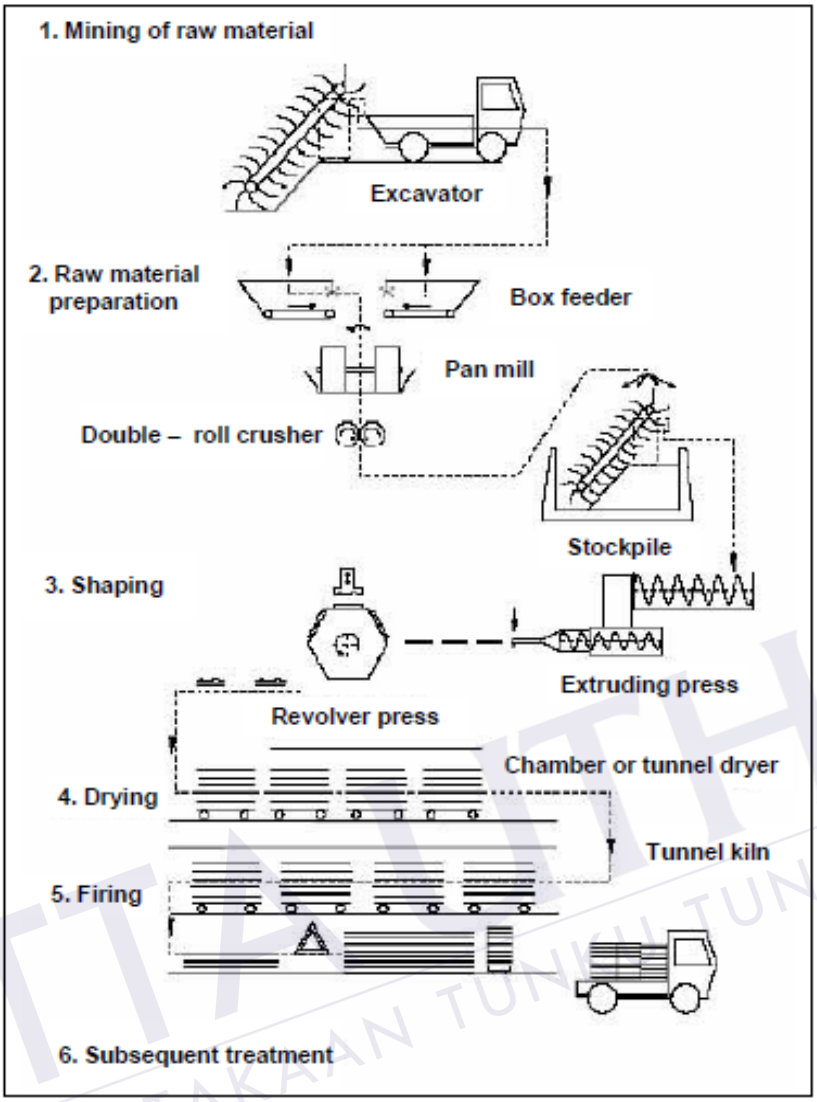

Figure 2.8: Schematic view of clay roof tiles manufacturing process (European Commission, 2007)

2.6.1.2 Raw Materials

Formulas (or bodies) of clay based ceramics may consists of one single clay or more clays, mixed with mineral modifiers which so called non-plastics such as powdered quartz and feldspar. Clays have layered structure. The layers are built from tetrahedral sheets in which a silicon atom is surrounded by four oxygen atoms and octahedral sheets in which a metal like aluminium or magnesium is surrounded by eight oxygen atoms. The tetrahedral and octahedral sheets are fused together by sharing oxygen atoms. Unshared oxygen atoms are present in hydroxyl form. Two main arrangements of tetrahedral and octahedral sheets fused together make the structure of clay (Olad, 2008).

Clay is mostly used for the roof tiles manufacturing. Organic and inorganic additive can be added in order to obtain a greater pore volume. In this roof tile industry, metallic oxides such as MnO_2, Fe_2O_3 and mineral such as $CaCO_3$ normally added to obtain the desired colour and/or to enhance porosity of the finished product. These additives are added just before pressing process or in the raw material preparation either in solid or liquid form (European Commission, 2007). Table 2.5 below indicates the basic ingredients for clay roof tile.

Table 2.5: Roofing tile constituents (Crowe, 2005)

Constituent	*Kg/m^2 (lb/square)**	*Mass Fraction*
Asphalt	0.57 (12)	45 %
Organic Felt	0.51 (10)	10 %
Limestone	0.13 (2.6)	5 %
Sand	0.06 (1.3)	40 %
Total	1.27 (25.9)	100 %

*One square is equivalent to 9.29 m^2 (100 ft^2)

2.7 Waste recycling

Increasing energy prices and depletion of natural resources are becoming pressing realities that need novel solutions. The availability of resources drawn directly from nature is decreasing and thus viable alternatives must be considered especially through recycling (Alami,2013). Most sectors of the ceramic industry recycle materials such as off-cuts, trimmings and substandard articles back to the raw material preparation stage. Fired goods which rejected during quality outgoing inspection due to process reject, firing issue etc. may also be recycled within the factories usually after crushing. Even if 'fired process loss' recycling is not suited to the process from which it originated, it may find an outlet in a different ceramic process. For clay roof tiles, recovery and reuse is widely practiced especially for bricks which is the demand is high. Besides, some of finely roof tiles also found use as partial aggregates in concrete or as a filler in asphalt for road construction (European Commission, 2007).

A study conducted by Herbudiman and Saptaji (2013) had justified two major reasons to select traditional roof- tiles powder as reinforced component in self compacting concrete. It is because the availability of the material which easy to get and directly gives positive impact to the environmentby recycling. The CRTW has silica content then potentially could be used as pozzolanic material. As pozzolanic material, CRTW could be a good binder and also a good filler agents. With suitable trial mix composition, traditional roof-tile powder could improve quality and flowability of self compacting concrete.

Most studies have confirmed the possibility to use ceramic waste in concrete, however due to different properties of such aggregates, parameters of obtained concrete types also differ. In general, substitution of traditional natural aggregates with red ceramics waste adversely affects concrete strength parameters. The higher the content of traditional aggregate substitued by red ceramic waste, the lower the concrete strength. Studies on concrete containing sanitary and technical ceramic waste have confirmed advantages brought about by such aggregates. Study results have shown that strength of concrete containing from 3% to 9% of coarse aggregates of ceramic sanitaryware waste was higher by 2% to 8% than that of concrete without this admixture. If more ceramic admixture was used the results were better (Halicka et al., 2013).

Extensive research and development work towards exploring new ingedients are required for producing sustainable and environment-friendly construction materials. The present study investigates the potential use of various solid wastes including ceramic wastes in the production of construction materials. Table 2.6 summarized several examples on the use of solid waste based construction materials in real construction (Safiuddin et al., 2010).

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